

# Violation of Ioffe-Regel condition but saturation of resistivity of the high $T_c$ cuprates

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We demonstrate that the resistivity data of a number of high  $T_c$  cuprates, in particular  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ , are consistent with resistivity saturation, although the Ioffe-Regel condition is strongly violated. By using the f-sum rule together with calculations of the kinetic energy in the  $t - J$  model, we show that the saturation resistivity is unusually large. This is related to the strong reduction of the kinetic energy due to strong correlation effects. The fulfilment of the Ioffe-Regel condition for conventional transition metal compounds is found to be somewhat accidental.

Some strongly correlated metals [1–4], in particular many high  $T_c$  cuprates [5–14], show an exceptionally large resistivity,  $\rho$ , at high temperatures  $T$ . Thus  $\rho$  can reach values of several  $\text{m}\Omega\text{cm}$ . This is in strong contrast to almost all other metals. Typically, a metal with a very large resistivity shows resistivity saturation [15]. Thus when  $\rho$  reaches values of the order  $0.1 \text{ m}\Omega\text{cm}$ , the slope of  $\rho(T)$  is typically reduced substantially, although not necessarily to zero. This happens when the apparent mean free path  $l$  becomes comparable to the separation,  $d$ , of two atoms, the Ioffe-Regel condition [16]. This kind of behavior has been observed for many metals and it used to be considered a universal behavior [17].

The metals mentioned above and the alkali-doped fullerenes [18] are apparent exceptions to this saturation behavior, with the cuprates forming a particular challenge [19,20]. The deviation from the “universal” behavior can be illustrated by considering Ioffe-Regel condition for  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ . We assume a “large” Fermi surface of cylindrical shape, containing  $1 - x$  electrons (or  $1 + x$  holes) [21]. Assuming that  $l = a$ , where  $a$  is the lattice parameter in the  $\text{CuO}_2$  plane, we obtain the saturation resistivity

$$\rho_{\text{sat}} = \frac{0.7}{\sqrt{1 \pm x}} \text{m}\Omega\text{cm}. \quad (1)$$

If we instead assume a “small” Fermi surface,  $\sqrt{1 \pm x}$  in the denominator is replaced by  $\sqrt{x}$ . In either case, the experimental resistivity for small  $x$  is much larger than the saturation resistivity above, supporting the conclusion that there is no saturation in these systems.

Here we demonstrate that the experimental data for the strongly correlated high  $T_c$  cuprates are, nevertheless, consistent with saturation. The saturation resistivity can, however, be much larger than the Ioffe-Regel value. This is not entirely surprising, since the Ioffe-Regel condition is based on a semiclassical picture, which is not valid when  $l \sim d$ . The good agreement between the Ioffe-Regel condition and the saturation for weakly correlated transition metal compounds is therefore somewhat accidental. The large saturation resistivity is shown to be due to a large reduction of the kinetic energy, due to strong correlation effects. Since the alkali-doped fullerenes appear to lack saturation [22], this puts these compounds in a special class, different from the cuprates.

To discuss resistivity saturation, we use the f-sum rule in a form appropriate for the models discussed here (only nearest neighbor hopping and no on-site matrix elements of the current operator) [23,24]

$$\frac{2}{\pi} \int_0^\infty \sigma(\omega) d\omega = -\frac{1}{2} \frac{d^2 e^2}{N \Omega \hbar^2} \langle T_K \rangle, \quad (2)$$

where  $\sigma(\omega)$  is the optical conductivity,  $d$  is the separation of the sites,  $\Omega$  is the volume of a unit cell,  $N$  is the number of unit cells and  $\langle T_K \rangle$  is the expectation value of the kinetic energy operator. The prefactor  $1/2$  refers to the two-dimensional case and is replaced by  $1/3$  for the three-dimensional case. To obtain an approximate upper limit to the resistivity, we assume that the (Drude) peak at  $\omega = 0$  has been smeared out and that  $\sigma(\omega)$  is a smooth function. The removal of the  $\omega = 0$  peak may be due to any scattering mechanism, electron-phonon [20], electron-electron or impurity scattering and, depending on the system, it may or may not happen at large values of  $T$ . We emphasize that our theory below does not depend on any particular scattering mechanism. We furthermore assume that  $\sigma(\omega) = 0$  for  $|\omega| > W$ , where  $W$  is the one-particle band width. This should be a good approximation, since  $|\omega| > W$  would involve multiple electron-hole pair excitations and have a small weight. Explicit calculations for a model of transition metal compounds and for the  $t - J$  model confirm this. If  $\sigma(\omega)$  had a box shape,  $\sigma(\omega) \equiv \sigma(0)$  for  $0 \leq |\omega| \leq W$ , the integral over  $\sigma(\omega)$  would be  $\sigma(0)W$  and we could write

$$\sigma(\omega = 0) = \frac{\gamma \hbar}{W} \int_0^\infty \sigma(\omega) d\omega, \quad (3)$$

with  $\gamma = 1$ . For a more realistic shape of  $\sigma(\omega)$ , where  $\sigma(\omega)$  has a maximum for  $\omega = 0$ , it follows that  $\gamma > 1$ . Following the considerations for a model of transition metal compounds with electron-phonon scattering [25], we may set  $\gamma = 1.9$ . In the  $t - J$  model, considering electron-electron scattering, we find that  $\sigma(\omega)$  is more concentrated to small  $\omega$ , giving a larger value of  $\gamma$ . If the electrons would tend to localize,  $\gamma$  could be much smaller than unity. This should, however, not happen at the large values of  $T$  considered here. Measurements [26] of  $\sigma(\omega)$  for  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  at large  $T$  show a similar

behavior as assumed in Eq. (3), apart from some sharp phonon structures at finite  $\omega$ .

We first consider noninteracting electrons in a band with the orbital degeneracy  $n$ . The scattering is assumed to be due to the electron-phonon interaction. For  $T \ll W$  and neglecting the influence of the electron-phonon coupling on the kinetic energy,

$$\langle T_K \rangle = 2n \int_{-W/2}^{\mu} \varepsilon N(\varepsilon) d\varepsilon \equiv -2n\alpha W N, \quad (4)$$

where  $N(\varepsilon)$  is the density of states per orbital and spin,  $\mu$  is the chemical potential and  $\alpha$  depends on the shape  $N(\varepsilon)$  and the filling. For semi-elliptical  $N(\varepsilon)$  and filling 0.4,  $\alpha = 0.10$ . This result is relatively independent of the specific shape of  $N(\varepsilon)$ , and using, e.g., a constant  $N(\varepsilon)$  leads to  $\alpha = 0.12$ . For a three-dimensional system we then obtain

$$\sigma(0) = \frac{\pi n}{3} \alpha \gamma \frac{d^3}{\Omega} \frac{e^2}{\hbar d}, \quad (5)$$

which provides an approximate upper limit to the resistivity for  $T \ll W$ . Saturation happens if  $\rho(T)$  grows so rapidly that this approximate upper limit is approached for experimentally accessible values of  $T$ . This happens for some transition metal compounds, with the A15 compounds, such as  $\text{Nb}_3\text{Sb}$ , being particularly striking examples. The important orbitals in  $\text{Nb}_3\text{Sb}$  are the Nb  $d$ -orbitals, leading to  $n = 5$ . Considering the A15 lattice of  $\text{Nb}_3\text{Sb}$ , we obtain the saturation resistivity  $0.14 \text{ m}\Omega\text{cm}$ , which compares well with the experimental saturation resistivity of about  $0.15 \text{ m}\Omega\text{cm}$  [15]. This illustrates the usefulness of this approach for obtaining a saturation resistivity.

We next consider the strongly correlated cuprates. To calculate the kinetic energy  $\langle T_K \rangle$ , we need an appropriate model for these systems. The transport properties should mainly be determined by the Cu-O antibonding band of Cu  $x^2 - y^2$  and O  $2p$  character. Thus we consider one orbital per site and the orbital degeneracy  $n = 1$ . These orbitals are put on a two-dimensional square lattice, and we consider a layered system with the lattice parameters  $(a, a, 2c)$ . We further assume that the Coulomb interaction  $U$  between two electrons on the same site is so large that double occupancy of a site can be neglected. This can be described by the  $t - J$  model [27]. The properties of this model have been studied extensively by Jaklic and Prelovsek [24]. The system has the hole doping  $x$  ( $> 0$ ). We emphasize that the  $t - J$  model is only used to estimate  $\langle T_K \rangle$ . We do not exclude other scattering mechanisms; we just neglect their contribution to the kinetic energy.

For simplicity, we first consider  $J = 0$ . By expanding in  $1/T$ , we obtain the large  $T$  limit of the kinetic energy as

$$\langle T_K \rangle = \frac{\text{Tr } T_K e^{-H/T}}{\text{Tr } e^{-H/T}} \approx -\frac{1}{T} \left[ \frac{\text{Tr } T_K^2}{\text{Tr } 1} \right]_{\text{no double occ}}, \quad (6)$$

where the right hand side is evaluated for states without double occupancy. Since the expression contains  $T_K^2$ , this involves a hole hopping to a neighboring site and back. In the  $t - J$  model this is only possible if the neighboring site has no holes. The probability for having a hole or no hole on a given site is  $x$  and  $(1 - x)$ , respectively. The probability for hopping between two nearest neighbor sites is then approximately  $x(1 - x)$ . Summing over the four nearest neighbors, this gives the large  $T$  limit

$$\langle T_K \rangle = -4Nt^2 \frac{x(1 - x)}{T}. \quad (7)$$

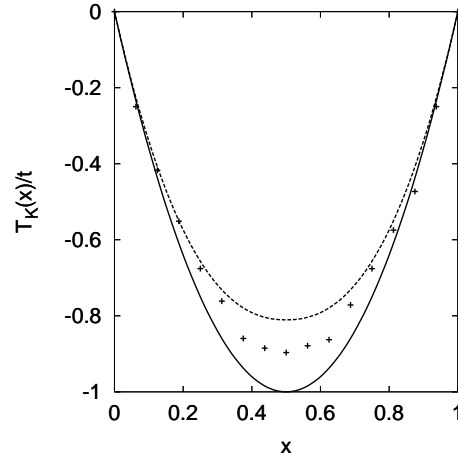


FIG. 1. The kinetic energy  $T_K(x)$  as a function of doping  $x$  in the  $t - J$  model for  $J = 0$ . The crosses show the result of exact diagonalization for  $4 \times 4$  two-dimensional cluster [30], the full curve Eq. (8) and the broken curve the result for spinless fermions [28,29]. The figure illustrates the approximate  $x(1 - x)$  behavior of the kinetic energy.

Using similar arguments, we can obtain a rough estimate for the  $T = 0$  kinetic energy

$$[\langle T_K \rangle]_{T=0} = -4tx(1 - x)N. \quad (8)$$

This result is shown by the full curve in Fig. 1. A similar result, but with a smaller coefficient, can be obtained variationally, by assuming that the holes behave as spinless fermions in a ferromagnetic background (broken curve in Fig. 1) [28,29]. These results can be compared with results from an exact diagonalization [30] of a  $4 \times 4$   $t - J$  model with  $J = 0$  (crosses in Fig. 1). The figure illustrates the approximate  $x(1 - x)$  dependence of the kinetic energy. In the following we use the  $t - J$  model with  $J/t = 0.3$  and  $t = 0.4 \text{ eV}$  [24]. We solve the model, using the finite  $T$  exact diagonalization technique of Jaklic and Prelovsek [31]. We consider a finite cluster of  $4 \times 4$  atoms and use the grand canonical ensemble. The resulting numerical results for the kinetic energy for small values of

$T$  and  $x$  can be approximately expressed in the form of Eq. (8), but with the prefactor 4 reduced to about 3.4. For the values of  $T$  of interest here, the  $T$  dependence of  $\langle T_K \rangle$  is weak, and it is neglected in the following. If the system has a strong electron-phonon interaction, the kinetic energy should be somewhat further reduced.

Combining Eqs. (2, 8), we predict that the right hand-side of Eq. (2) should be proportional to  $x$ , for small  $x$ . The effective number of carriers per Cu atom  $N_{eff}(x)$  can be estimated integrating the optical conductivity up to  $\omega_c = 1.2$  eV, as suggested by Yamada *et al.* [32]. This also includes some contribution from interband transitions, and Yamada *et al.* [32] therefore subtracted  $N_{eff}(0)$ , which should contain only interband transitions. They found that  $N_{eff}(x) - N_{eff}(0)$  is indeed approximately proportional to  $x$ , up to  $x = 0.12$ . Based on the absolute numbers by Uchida *et al.* [33], we find that our factor of proportionality in Eqs. (2, 8) agrees with experiment to within about 20-30 %. Due to the problems of removing the interband transitions, this should be a good agreement.

We consider conduction in the ab-plane of a cuprate and assume that the  $\omega = 0$  peak is gone. Insertion of Eq. (8), but with the prefactor 3.4, in Eqs. (2, 3) gives an approximate upper limit to the resistivity for small and intermediate values of  $T$

$$\rho = \frac{0.4}{x(1-x)} \text{ m}\Omega\text{cm}, \quad (9)$$

where we have used distance  $c = 6.4$  Å between the CuO<sub>2</sub> planes. Inserting the calculated kinetic energy for the  $t - J$  model would lead to a somewhat larger resistivity for small  $x$  and it would introduce a weak  $T$  dependence. The saturation resistivity in Eq. (9) is much larger than Eq. (1) for small  $x$ . It illustrates that one should expect saturation at a substantially larger resistivity than predicted by the Ioffe-Regel condition. The saturation resistivity in Eq. (9) is also much larger than the result derived [25] for a model of weakly correlated transition metal compounds

$$\rho \sim \frac{0.2d}{n} \approx \frac{0.5}{n}, \quad (10)$$

where  $d$  is expressed in Å. This is partly due to the degeneracy being just  $n = 1$  for the  $t - J$  model but  $n = 5$  for our model of the weakly correlated transition metal compounds. Furthermore, the strong correlation drastically reduces the kinetic energy in the cuprates, which shows up as the factor  $x(1-x)$  in the denominator of Eq. (9). Finally, the large value of the lattice parameter  $c$  also increases  $\rho$  for the cuprates.

We now consider the  $T$  dependent resistivity for different doping. The experimental results of Takagi *et al.* [6] are shown in Fig. 2. For  $T \leq 300$  K, more recent results have been obtained by Ando *et al.* [7]. Their resistivities are qualitatively similar but generally smaller

than the results in Fig. 2. The results by Takagi *et al.* [6] are compared with the saturation resistivity in Eq. (9). For all values of  $x$ , the experimental resistivity is smaller than the saturation resistivity predicted here. The experimental results do therefore not demonstrate absence of saturation. Actually, for  $x = 0.04$  and  $x = 0.07$  the data show signs of saturation for values not much smaller than the expected saturation resistivities, but at much larger values than the saturation resistivity predicted by the Ioffe-Regel criterion (Eq. (1)) (indicated by an arrow). This shows that the Ioffe-Regel criterion is invalid for these systems. It is interesting to plot  $x\rho(T)$  [34]. In such a plot the saturation resistivity becomes almost independent of  $x$ . Indeed the curves of  $x\rho(T)$  for  $x = 0.04$  and  $x = 0.07$  fall almost on top of each other, suggesting that close to saturation there is a scaling of  $\rho(T)$  by  $1/x$ , as implied by the arguments above. For  $x = 0.15$  and  $x = 0.34$  one would have to study much higher (and experimentally inaccessible) values of  $T$  to determine whether or not there is saturation.

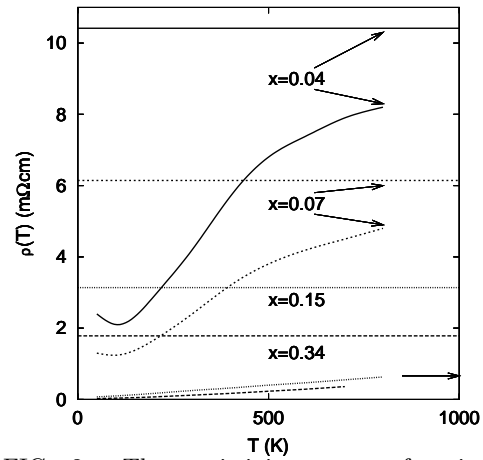


FIG. 2. The resistivity as a function of  $T$  for  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  [6] and the saturation resistivity according Eq. (9) for  $x = 0.04$  (full curve)  $x = 0.07$  (broken curve)  $x = 0.15$  (dotted curve) and  $x = 0.34$  (chain curve). The horizontal arrow shows the saturation resistivity expected from the Ioffe-Regel condition. The figure illustrates that there are signs of saturation for small  $x$  at roughly those resistivities where saturation is expected. For larger  $x$ , much larger  $T$  would have to be considered to test whether there is saturation.

$\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  has a structural transition from an orthorhombic to a tetragonal phase. This transition happens, however, at a somewhat lower temperature [32] than the break in the slopes of the curves for  $x = 0.04$  and  $x = 0.07$  in Fig. 2.

We have also considered the resistivity of  $\text{YBa}_2\text{Cu}_3\text{O}_{6+\delta}$  [8],  $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_{4-y}$  [9],  $\text{Bi}_2\text{Sr}_2\text{Ca}_{1-x}\text{Y}_x\text{Cu}_2\text{O}_{8+y}$  [10,11],  $\text{Tl}_2\text{Ba}_2\text{CuO}_{6+\delta}$  [12] and  $\text{HgBa}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_{2n+2+x}$  [13]. In all these cases

we find that the experimental resistivity does not exceed the expected saturation resistivity. In the case of  $\text{Bi}_2\text{Sr}_2\text{Ca}_{1-x}\text{Y}_x\text{Cu}_2\text{O}_{8+y}$  saturation has been reported [11]. This provides further support for our theory.

Above we have provided an upper limit to the resistivity based on the f-sum rule and the calculated kinetic energy and without specifying a scattering mechanism. We have also calculated the resistivity of the  $t-J$  model directly, although the results are not very accurate for the small clusters considered here. For  $x = 0.15$ , the calculated resistivity is of the right order of magnitude, while for  $x = 0.07$  and  $x = 0.04$  it is substantially smaller than the experimental results in Fig. 2. This suggests that there is an additional scattering mechanism, beyond the electron-electron scattering included explicitly in the  $t-J$  model, which becomes important for  $x = 0.04$  and  $x = 0.07$ . This additional mechanism apparently makes the resistivity increase so rapidly for small  $T$  that the approximate upper limit (9) is approached and the resistivity shows sign of saturation. The estimate of the upper limit (9) is nevertheless correct, unless the additional scattering mechanism influences the kinetic energy in a substantial way.

Above we have discussed the situation when  $T$  is much smaller than the band width, which applies for values of  $T$  which can be reached experimentally. The kinetic energy  $T_K$  then has a weak  $T$  dependence, leading to an upper limit for the resistivity which is only weakly  $T$  dependent. As can be seen from Eq. (7), however, the  $T$  dependence becomes strong for very large  $T$  ( $> W/4$ ). Inserting Eq. (7) into Eqs. (2,3) then leads to an upper limit for  $\rho(T)$  which grows linearly with  $T$ . A similar result would be found even for noninteracting electrons scattered by impurities, i.e., for a  $T$ -independent mechanism, where the  $T$  dependence can then be traced back to the  $T$  dependence of the Fermi functions. This illustrates that “saturation” does not necessarily imply that the resistivity becomes  $T$  independent, just that its slope is reduced substantially at large  $T$  [17]. The effects discussed in this paragraph, however, refer to very large values of  $T$ , of the order of 10000 K. They are therefore only of academic interest, unless the band width is very small.

To summarize, we have shown that the experimental resistivity data for a number of cuprates are consistent with resistivity saturation. The saturation resistivity is, however, much larger than for, e.g., A15 compounds, due to the strong suppression of the kinetic energy in the strongly correlated cuprates. The Ioffe-Regel condition is therefore violated for a many of these systems.

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